Results of UREX Hot Demonstration

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Tracy Rudisill

Westinghouse Savannah River Company

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Background

- Transmutation of Waste is being Developed by the Advanced Fuel Cycle Initiative
  - Address Disposal of Commercial Nuclear Fuel
  - Improve Performance of the Geologic Repository
- Transmutation Program will Require Separation of Reactor Fuel into . . .
  - TRU Product Stream which is Converted to Fuel and Transmuted by Fissioning to Generate Power
  - Separate ^{99}Tc and ^{129}I Streams which are Converted to Targets and Transmuted to Short-lived Nuclides
  - Uranium Product that Meets Criteria for Class C LLW

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UREX Process

- The PUREX Process is a Mature Solvent Extraction Process for Irradiated Fuel Designed to Recover Pu and U
- A Variation of the PUREX Process was Conceived to Treat Large Quantities of Irradiated Spent Fuel and to Provide High Selectivity
  - PUREX was Modified so Only U and Tc are Extracted and the TRU Isotopes (Np, Pu, Am, and Cm) are Rejected with the Fission Products
  - This Uranium Extraction Process is Called UREX

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UREX Process Goals

- Recovery Efficiencies
  - Recover >99.9% of the U and >95% of the Tc
  - Reject >99.9% of the TRU Isotopes to the Acid Waste (Raffinate)
  - U Product Meets Requirements for Class C LLW
    - Fission Products (Primarily Cs and Sr)
    - TRU Isotopes (<100 ηCi/g)
- Waste Minimization
  - Conversion of All Chemicals to Gases During Subsequent Processing
  - Acetohydroxamic Acid (AHA) Used to Prevent the Extraction of Pu and Np

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Chemistry of AHA in the UREX Process

- AHA is an Analogue of Hydroxylamine
  - Hydroxylamine Nitrate is used as a Reductant in Nuclear Processing
- AHA Serves as Both a Reductant and a Complexant in the UREX Process
  - Reduces Np (VI) to Inextractable Np (V)
  - Prevents Extraction by Complexing Pu (IV) and Np (IV)
- AHA Readily Decomposes to Gaseous Products During Waste Evaporation
- AHA Hydrolyzes in Acidic Solutions

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Hot UREX Demonstration

- A "Hot" Demonstration of the UREX Process was Performed in the SRTC Shielded Cells Facility Using Spent Fuel from the Dresden Reactor
- UREX Demonstration Performed with Centrifugal Contactors Previously Used to Demonstrate the Extraction of Cs from SRS Waste

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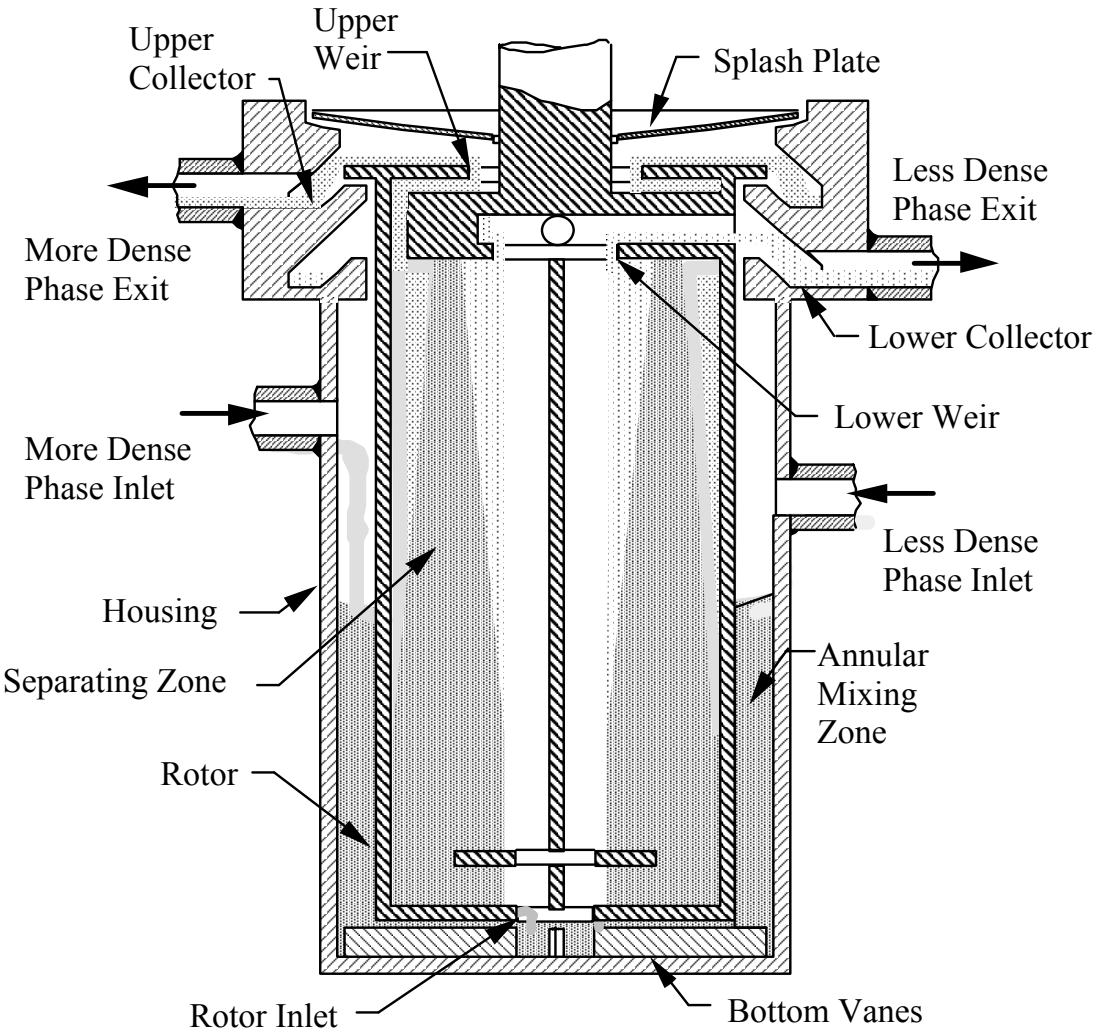
Centrifugal Contactor Schematic



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Dresden Reactor Fuel

- Boiling-Water Reactor Near Morris, IL
- Fuel Discharged on September 1, 1975
- Fuel Burnup 23,480 MWD/MT
- Fuel Contained 4 kg of U and 29 g of Pu

Fuel Dissolution

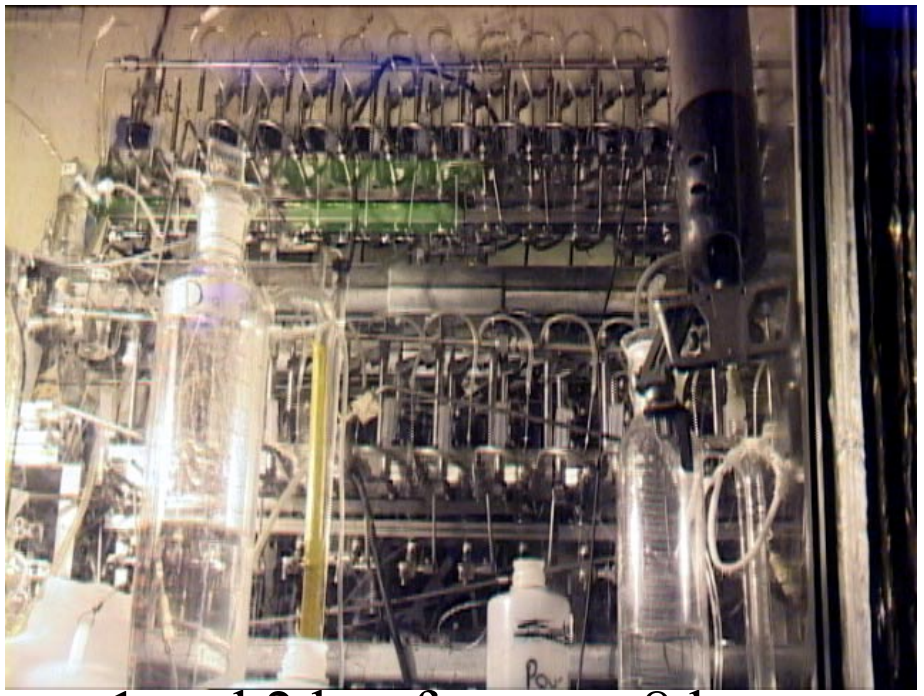
- Fuel Segments were Dissolved using a 6 L Glass Dissolver and Heating Mantle
  - Stainless Steel Basket used to Contain the Fuel
  - Offgas was Condensed with Water
- Two Step Dissolution Performed using HNO_3 at 90ºC
  - Initial Dissolution Performed in 1-4M HNO_3
  - Following Temperature Spike 10M HNO_3 was Added to Dissolver to Complete Dissolution
- Hulls were Leached with 4M HNO_3 at 90 ºC for 2-6 h

Flow Rate Control for Hot UREX Demonstration

- Positive Displacement and Peristaltic Feed Pumps were Manually Controlled
- 2 or 4 L Graduated Cylinders with Attached Burette used for Flow Measurements
- Goal - Measure Flow Rates Once per Hour or As Needed

Hot Demonstration Tests

- 3 Hot Tests Performed
  - Two 6 h Tests
  - One 30 h Test with Changes in Flow Conditions
- 30 h Test
  - Test 3A Performed at Same Flow Conditions as Tests 1 and 2 but for an 8 h Duration
  - Test 3B Performed at Flow Conditions which Generated More Concentrated U and Tc Products



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Analytical Methods

Analysis	Method
U	Spectrophotometrically (1-10 g/L U)
	ICP-MS (Trace Concentrations)
	Laser Fluorescence (Trace Concentrations)
^{90}Sr	Beta Counting (Ion Exchange Chromatography)
Pu	Alpha Pulse Height Analysis (TTA Extraction)
Tc	Beta Counting (Ion Exchange Separation)
Gamma Emitters	Gamma Pulse Height Analysis
Other Elements	ICP-ES or ICP-MS

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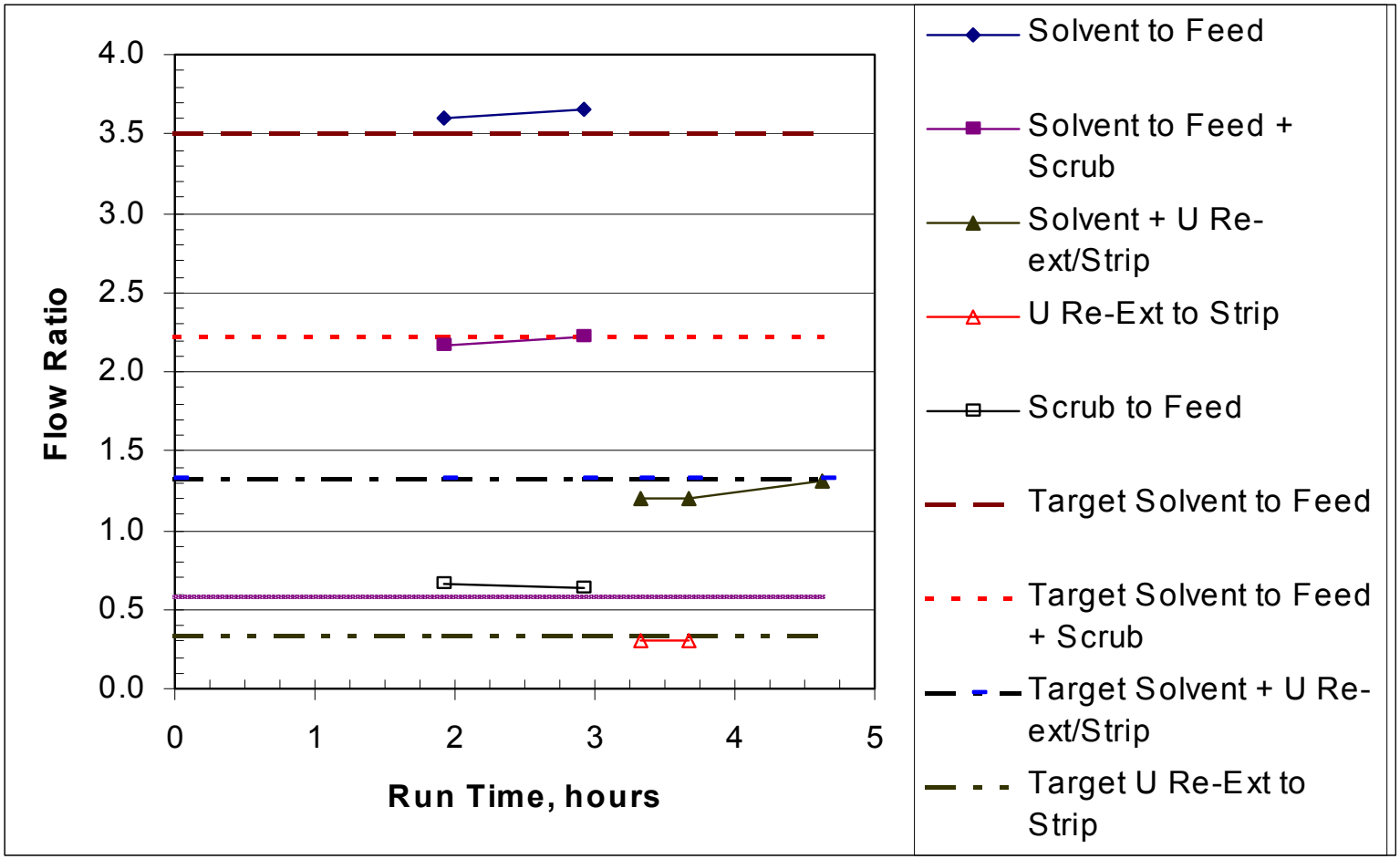
Flow Rate Ratios

UREX Test 2



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Analytical Results

UREX Test 2

Stream	Time (hr)	U (g/L)	Pu (d/m/mL)	Am (d/m/mL)	Tc (d/m/mL)	Cs (d/m/mL)	Sr (d/m/mL)
U Product	0	3.9	4.12E+04	1.16E+04	2.11E+03	7.90E+04	1.02E+04
	1	51.2	4.43E+03	2.85E+03	2.72E+03	6.05E+03	3.84E+03
	3	54.5	6.01E+03	NA	1.15E+04	8.98E+03	3.85E+03
	4	53.7	1.18E+04	1.53E+04	2.07E+03	1.17E+04	3.85E+03
	6	55.1	3.02E+03	5.07E+03	1.29E+03	1.67E+04	5.00E+03
Tc Product	0	0.0045	NA	NA	2.25E+04	1.53E+05	2.10E+04
	2	0.0045	2.41E+03	NA	1.58E+06	1.28E+05	1.46E+04
	3	0.0045	2.31E+04	1.85E+04	1.82E+06	4.48E+05	2.30E+05
	4	0.0045	8.41E+03	5.00E+03	1.81E+06	3.34E+05	1.02E+05
	6	0.000079	4.67E+03	7.53E+03	1.45E+06	3.93E+05	1.11E+05
Waste Raffinate	1	NA	6.72E+08	8.79E+08	1.64E+05	1.77E+10	NA
	2	NA	8.55E+08	6.57E+08	5.86E+05	1.26E+10	9.78E+9
	3	NA	1.04E+09	NA	5.74E+05	1.93E+10	1.30E+10
	4	0.0026	7.57E+08	NA	1.46E+05	1.68E+10	1.94E+10
	6	0.0297	7.97E+08	7.50E+08	2.91E+05	1.89E+10	1.47E+10

NA - No Analysis

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Material Balance Results

UREX Test 2

Stream	Time (hr)	U (%)	Pu (%)	Am (%)	Tc (%)	Cs (%)	Sr (%)
U Product	0	6	0.0187	0.00513	<0.17	0.00181	0.000224
	1	84	0.00201	0.00126	<0.21	0.000138	0.000084
	3	90	0.00276	NA	<0.91	0.000208	0.000085
	4	89	0.00542	0.00682	<0.16	0.000271	0.000085
	6	91	0.00138	0.00226	<0.10	0.000386	0.000111
Tc Product	0	<0.0061	NA	NA	1.4	0.00286	0.000376
	2	<0.0061	0.000895	NA	101	0.00239	0.000243
	3	<0.0061	0.00868	0.00676	118	0.00848	0.00388
	4	<0.0061	0.00314	0.00182	116	0.00629	0.00171
	6	0.000079	0.00161	0.00252	86	0.00682	0.00171
Waste Raffinate	1	NA	103	131	4	136	NA
	2	NA	131	98	15	97	72
	3	NA	157	NA	15	146	94
	4	0.0014	114	NA	4	127	141
	6	0.0161	120	110	8	143	107

NA - No Analysis

Contamination Levels

UREX Uranium Product Stream

Nuclides	LLW Limit, Ci/m^3			UREX Test Results[(1)]			
	Class A	Class B	Class C	Test 1	Test 2	Test 3A	Test 3B
^{99}Tc			3	0.34	0.03	NA	NA
^{129}I			0.08	NM	NM	NM	NM
Nuclides (half-life <5yr)	700			0.05	ND	ND	ND
^{90}Sr	0.04	150	7000	0.22	0.12	<0.07	<0.07
^{137}Cs	1	44	4600	0.47	0.41	0.04	0.03-0.11
TRU Isotopes (half-life >5yr)			100 ηCi/g	<296	65	16	29-153
^{241}Pu			3500 ηCi/g	1420	214	71	147-1020
^{242}Cm			20000 ηCi/g	ND	ND	ND	ND

(1) Assumes density of bulk UO_3 is half the crystal density or 3.645 g/cm$_3$

NA - Not Available

ND - Not Detected

NM - Not Measured

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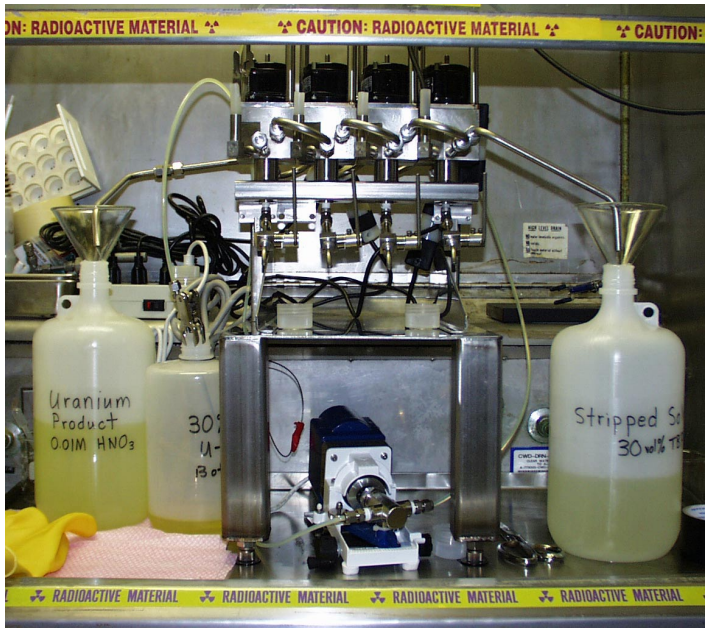
Uranium Stripping

- Number of Stages in Cell was Insufficient to Strip U from the Solvent
- 4 Centrifugal Contactor Stages Set-up in Hood
  - U Stripped from Solvent with 0.01M Nitric Acid



- Whole Body Dose Rate of Bottles Containing U-Loaded Solvent was <1 mRad/hr
  - Low Dose Rate Demonstrates Good Fission Product Decontamination Achieved by UREX Process
- UO_3 Produced by Evaporation, Precipitation of $(NH_4)_2U_2O_7$, and Calcination

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UO_3 Product

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Conclusions

- Demonstration of UREX Process at Baseline Conditions Showed All Goals for Recovery and Decontamination are Achievable
  - U Product was Class C (or Lower) LLW
  - Recovery of >99.9% U and >95% Tc
  - Rejection of >99.9% of Pu to the Raffinate
- Tc Losses to U Stream were 1.2% in 1st Test and 0.1% in 2nd Test
- Loss of Pu and Other Actinides to the Tc and U Product Streams was <0.1% in All Tests

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Flowsheet for UREX Test 3B